(1)

PCT

WORLD INTELLECTUAL PROPERTY ORGANIZATION
International Bureau

INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classificati n 6 : H01M 10/04, 6/14	A1	(11) International Publication Number: WO 96/01506 (43) International Publication Date: 18 January 1996 (18.01.96)

(21) International Application Number: PCT/US95/07560

(22) International Filing Date: 13 June 1995 (13.06.95)

(30) Priority Data:
08/270,620 5 July 1994 (05.07.94) US

(71) Applicant: MOTOROLA INC. [US/US]; 1303 East Algonquin Road, Schaumburg, IL 60196 (US).

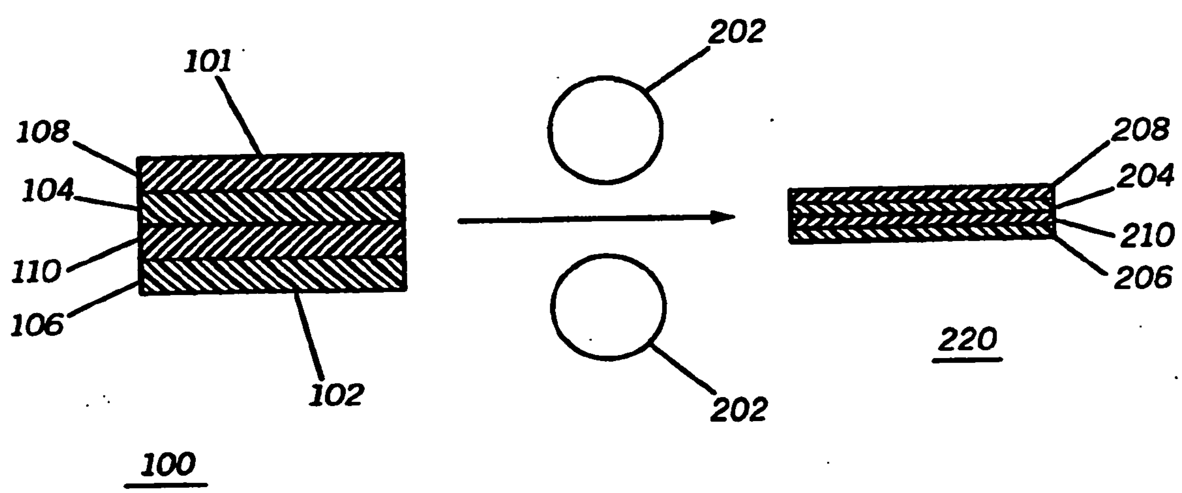
(72) Inventor: OLIVERIA, Manuel, P.; 5851 Holmberg Road #2624, Parkland, FL 33067 (US).

~~(74) Agents: FULLER, Andrew, S. et al.; Motorola Inc., Intellectual~~ Property Dept., 8000 West Sunrise Boulevard, Fort Lauderdale, FL 33322 (US).

(81) Designated States: AM, AT, AU, BB, BG, BR, BY, CA, CH, CN, CZ, DE, DK, EE, ES, FI, GB, GE, HU, JP, KE, KG, KP, KR, KZ, LK, LR, LT, LU, LV, MD, MG, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, TJ, TT, UA, UZ, VN, European patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, ML, MR, NE, SN, TD, TG), ARIPO patent (KE, MW, SD, SZ, UG).

Published
With international search report.

(54) Title: COMPOSITE BATTERY AND METHOD FOR FORMING SAME



(57) Abstract

A composite battery (600) and a method for forming the battery (600) are provided. The battery (600) is formed combining at least an electrode layer (108, 110) with an electrolyte layer (104, 106) into a stack (400) and by compressing the stack (400) to thin each layer.

FOR THE PURPOSES OF INFORMATION ONLY

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AT	Austria	GB	United Kingdom	MR	Mauritania
AU	Australia	GE	Georgia	MW	Malawi
BB	Barbados	GN	Guinea	NE	Niger
BE	Belgium	GR	Greece	NL	Netherlands
BF	Burkina Faso	HU	Hungary	NO	Norway
BG	Bulgaria	IE	Ireland	NZ	New Zealand
BJ	Benin	IT	Italy	PL	Poland
BR	Brazil	JP	Japan	PT	Portugal
BY	Belarus	KE	Kenya	RO	Romania
CA	Canada	KG	Kyrgystan	RU	Russian Federation
CF	Central African Republic	KP	Democratic People's Republic of Korea	SD	Sudan
CG	Congo	KR	Republic of Korea	SE	Sweden
CH	Switzerland	KZ	Kazakhstan	SI	Slovenia
CI	Côte d'Ivoire	LI	Liechtenstein	SK	Slovakia
CM	Cameroon	LK	Sri Lanka	SN	Senegal
CN	China	LU	Luxembourg	TD	Chad
CS	Czechoslovakia	LV	Latvia	TG	Togo
CZ	Czech Republic	MC	Monaco	TJ	Tajikistan
DE	Germany	MD	Republic of Moldova	TT	Trinidad and Tobago
DK	Denmark	MG	Madagascar	UA	Ukraine
ES	Spain	ML	Mali	US	United States of America
FI	Finland	MN	Mongolia	UZ	Uzbekistan
FR	France			VN	Viet Nam
GA	Gabon				

COMPOSITE BATTERY AND METHOD FOR FORMING SAME

Technical Field

This invention relates in general to composite batteries, and more particularly, to composite batteries using stacked electrochemical cells.

Background of the Invention

Conventional battery manufacturing techniques typically involve the fabrication of individual electrode and electrolyte layers which are combined to form electrochemical cells. Recently, cells have been formed using solid-state technology to provide a longer service life, relatively high energy densities, and higher discharge and recharge rates. For example, a typical cell may have a lithium anode, an oxide cathode, and a polymer electrolyte. Generally, the electrodes and electrolytes are formed using layers in a stacked arrangement for convenience in manufacturing. Additionally, various attempts have been made to increase charge densities, and discharge and recharge rates to produce a more flexible and efficient battery.

One known method of increasing charge density and discharge rates in a typical lithium solid-state battery is to reduce the thickness of individual layers of electrodes and electrolytes. However, using current manufacturing techniques, the reduction in layer thickness has limitations. For example, using extrusion or rolling, a lithium metal anode can achieve a thickness of 20-50 microns. A polymer film electrolyte can be formed with a thickness of 10-20 microns using a blown film or cast film process. An oxide cathode having a thickness of 50-100 microns can be achieved using a tape casting process. However, at these thicknesses, the individual layers are difficult to handle and may be expensive to use in manufacturing.

It is desirable to produce solid state electrochemical cells with very thin layers of electrodes and electrolytes, to shorten the separation between electrodes, and to achieve a higher electrolyte surface area. Current approaches to forming thin layers, such as vacuum deposition

techniques, or by combining thin layers as described above, result in high manufacturing costs. Therefore, it is desirable to have an electrochemical cell or a battery constructed therefrom, which utilizes a less expensive manufacturing process.

Brief Description of the Drawings

FIG. 1 is a cell stack in accordance with the present invention.

FIG. 2 is a process for compressing the cell stack, in accordance with the present invention.

FIG. 3 is a process for combining cell stacks to form a battery, in accordance with the present invention.

FIG. 4 is a staggered cell stack in accordance with the present invention.

FIG. 5 is a compressed staggered cell stack, in accordance with the present invention.

FIG. 6 is a composite battery made up of multiple staggered cell stacks, in accordance with the present invention.

Detailed Description of the Preferred Embodiment

While the specification concludes with claims defining the features of the invention that are regarded as novel, it is believed that the invention will be better understood from a consideration of the following description in conjunction with the drawing figures, in which like reference numerals are carried forward.

Generally, the present invention provides a composite battery and a method for forming the battery. The battery comprises stacked layers of electrodes and electrolytes which are compressed to provide substantially thin individual layers. In another embodiment, a staggered stack is used to provide electrical interconnection between multiple cell stacks, thereby facilitating the formation of a battery.

Referring now to FIG. 1, a cell stack 100 is shown in accordance with the present invention. The cell stack 100 includes layers of electrodes 108, 110 and electrolytes 104, 106 which are stacked together to form a composite structure 100 having first and second opposing surfaces 101, 102. Preferably, the cell stack 100 has an electrolyte layer disposed between electrode layers of opposing polarities. Thus, in the preferred embodiment, there is at least an anode layer 108, a cathode layer 110, and

an electrolyte layer 104 interposed between the anode and cathode layers 108, 110, thereby forming an electrochemical cell. Moreover, a second electrolyte layer 106 is formed as an outer layer to one of the electrode layers 110 to facilitate the combination of multiple stacks. The four layer structure 100 is laminated to hold the layers together. Although the cell stack 100 of the preferred embodiment has a particular configuration of electrode and electrolyte layers, numerous variations of layer combinations are possible while incorporating the concepts taught by the present invention.

In the present invention, the cell stack 100 is formed to have layers with specific mechanical properties. Each layer responds to a compressive ~~force applied perpendicularly to the first or second surface 101, 102 of the~~ stack, by extruding in a direction perpendicular to the force. Significantly, each layer should thin proportionally, while retaining its electrochemical properties. Preferably, each layer responds, substantially similarly, to thermal and mechanical forces applied to the cell stack 100. The dimensions of each layer can vary to account for differences in expected responses to compressive forces. The layers should maintain structural integrity after being compressed so that electrodes of opposing polarities remain separated from each other. In the preferred embodiment, the compositions of the electrolyte and electrode layers are chosen such that the layers have substantially similar responses to compressive forces applied to the cell stack 100.

The electrodes 108, 110 contain a solid active material ($LiNiO_2$ - cathode, Graphite - anode), in powdered form, in addition to an ionically conducting polymeric phase, which acts as a binder. Additionally, the cathode contains carbon black which increases the electrical conductivity. The composition of the cathode layer 110 comprises: 3.1% lithium imide, 13.5% copolymer of vinylidene fluoride (VDF) and tetrafluroethylene (TFE), commercially available as Elf Atochem Kynar 7201; 6.7% ethylene carbonate; 6.7% propylene carbonate; 62.0% lithium nickel oxide; 8.0% carbon black. The composition of the anode layer 108 comprises: 5.2% lithium imide; 22.4% copolymer of VDF and TFE; 11.2% ethylene carbonate; 11.2% propylene carbonate; 50.0% graphite, commercially available as Lonza, KS-15. The electrolyte layer 104, 106 includes an ionically conductive polymer, which is filled with an inert solid powder so that its mechanical properties are similar to the electrode layers. The

composition of the electrolyte comprises: 3.5% lithium imide; 15.0% copolymer of VDF and TFE; 7.5% ethylene carbonate; 7.5% propylene carbonate; 66.5% calcium oxide.

Referring to FIG. 2, a process for compressing a cell stack to achieve thin electrode and electrolyte layers is shown, in accordance with the present invention. The laminated cell stack 100 structure is compressed by applying a rolling process. Thus, the cell stack 100 is passed through rollers 202 to produce a thinner structure 220 having substantially thinner layers 204, 206, 208, 210. The compressed stack 220 can be repeatedly compressed to achieve even thinner structures. Note that the stack 100 undergoes significant compression to substantially reduce its thickness and should not be confused with slight deformations experienced during typical lamination processes. For example, a typical stack is fabricated with thick initial layers, i.e., greater than 100 microns. The thickness of the initial stack facilitates handling during the manufacturing process.

FIG. 3 shows a process for combining cell stacks, in accordance with the present invention. Multiple cell stacks 302, 304 are combined, and a rolling process applied to compress the combined stack to form a thinner structure 320. Cell stacks 220 processed in the first compression step (see FIG. 2) can be combined and compressed again to thin each layer even more, thereby forming more compact cell stacks having multiple anode, electrolyte, cathode combinations. Again, additional cell stacks can be further combined by cell stacks can be combined and the rolling process repeated to produce a composite structure having several substantially thin layers of electrodes and electrolytes. Moreover, by electrically coupling the anode and cathode layers, a high density battery can be formed which has high charge and discharge rates.

Referring to FIG. 4, a staggered cell stack 400 is shown in accordance with the present invention. The cell layers, i.e., the anode, cathode, and electrolyte layers 108, 110, 104, 106, are arranged to facilitate the electrical connection of multiple cell stacks to form a composite battery. In a preferred embodiment, the cell stack layers are arranged such that on a first end 415 of the stack 400, an end portion 409 of the cathode layer 110 is recessed behind end portions 405 of two electrolyte layers 104, 106, which are themselves aligned. The end portion 409 of the anode layer 108 on the first end 415 of the stack 400 extends beyond the end portion 405 of the electrolyte layers 104, 106. On a second

end 416 of the cell stack 400, the relationships between the end portions of the layers are reversed, such that an end portion 411 of the cathode layer 110 protrudes beyond the other layers 104, 106, 108. FIG. 5 shows a compressed staggered cell stack 500, in accordance with the present invention. The cell stack 500 is compressed using a rolling process similar to that described above. As a result, the electrolyte layers 104, 106 merge at a first end 515 of the cell stack 500. Correspondingly, the anode layer 108 has an extruding portion 509 at the first end 515 of the cell stack 500, while the cathode layer 110 has an extruding portion 511 at a second end 516 of the cell stack 500.

FIG. 6 is a composite battery 600 made up of multiple staggered cell stacks, in accordance with the present invention. By proper arrangement, electrical connections between cells may be formed simply by stacking the cell stacks appropriately. Multiple cell stacks 602, 604 are arranged such that the extruding portions 509, 609 of the anode layers from each cell are in electrical contact. Likewise, the extruding portions 511, 611 of the cathode layers of each cell are in electrical contact. The combination of cell stacks 600 are then compressed to further reduce its overall thickness. The process of compressing also causes the extruding portions of the anode and cathode layers 509, 609, 511, 611 to merge, respectively. Thus, a battery can be made efficiently and inexpensively using cells formed accordingly to the present invention.

The procedures for forming a composite battery, according to the present invention, are now summarized. A cell stack is provided having one or more electrode layers, and an electrolyte layer. Preferably, the cell stack comprises an electrolyte layer interposed between an anode layer and a cathode layer. Multiple cell stacks are combined to form a battery. In the preferred embodiment, the stack is formed by combining an anode layer, a cathode layer, and two electrolyte layers (one of which being disposed between the anode and cathode layers). Preferably the layers selected have substantially similar thermal and mechanical properties. Alternatively, the dimensions of the layers are selected such that compression of the layers will yield thinned layers of predictable dimensions. The cell stack is compressed, using a rolling process, to substantially thin each layer . In another embodiment the layers of the cell stack are arranged such that they have staggered end portions. The end portions are staggered such that when the cell stack is compressed,

layers of like compositions have portions that merge. Additionally, it is preferred that the cell layers are staggered such that combination of cell stacks have protruded portions of anode material at one end, and protruded portions of cathode at another end, such that when compressed these protruded portions are merged. Multiple staggered cell stacks are combined to establish electrical connections among the anode layers on the one hand, and electrical connections among the cathode layers on the other hand.

The composite battery and method for forming the battery taught by the present invention provides several advantages. Electrochemical cells suitable for forming high density batteries can be created with very thin cell layers. The thin cell layers enable the battery to provide high charge densities and high discharge and recharge rates. Additionally, the cells can be created by a relatively simple and low-cost manufacturing process. By achieving a thin layer electrolyte design, the performance specifications for the electrolyte can be relaxed, i.e., a poorer performing electrolyte can be used. One of the biggest challenges in developing a solid state polymer battery is attaining sufficient conductivity in a polymer. A cell constructed according to the present invention provides a short path length between the electrodes of opposing polarities by providing an electrolyte having a substantially thin structure. This enables electrolytes having a low conductivity to be used.

While the preferred embodiments of the invention have been illustrated and described, it will be clear that the invention is not so limited. Numerous modifications, changes, variations, substitutions and equivalents will occur to those skilled in the art without departing from the spirit and scope of the present invention as defined by the appended claims.

What is claimed is:

Claims

1. A method for forming a composite battery, comprising the steps of:

providing an electrode layer;
providing an electrolyte layer;
forming a first stack by combining the electrode layer and the electrolyte layer; and
compressing the first stack to thin each layer.

2. The method of claim 1, wherein the step of compressing the first stack, comprises the step of applying a rolling process to thin the layers of the first stack.

3. The method of claim 1, further comprising the steps of:
forming a second stack comprising an electrode layer and an electrolyte layer;
combining the first and second stacks; and
compressing the first and second stacks to thin each layer of the first and second stacks.

4. A method for forming a composite battery, comprising the steps of:

forming a stack, comprising the steps:

providing first and second electrode layers having opposing polarities;

providing first and second electrolyte layers;

disposing the first electrolyte layer between the electrode layers; and

disposing the first electrode layer between the electrolyte layers;

~~arranging the electrode and electrolyte layers to have staggered end~~ portions such that the first electrode layer has an end portion protruding at a first end of the stack, and the second electrode layer has an end portion protruding at a second end of the stack; and

compressing the stack to substantially thin each layer, to extrude a portion of the first and second electrode layers at the first and second ends of the stack, respectively, and to merge portions of the electrolyte layers.

5. The method of claim 4, further comprising the steps of:

forming a stack combination by combining multiple stacks ;

compressing the stack combination to electrically connect extruding portions of electrodes having similar polarities from the multiple stacks.

6. A method for forming a composite battery, comprising the steps of:

forming a cell stack, comprising the steps of:

providing an anode layer;

providing a cathode layer; and

providing an electrolyte layer interposed between the anode layer and the cathode layer; and

compressing the cell stack such that the anode layer, the cathode layer, and the electrolyte layer are thinned.

7. ~~The method of claim 6, wherein the steps of providing an anode~~ layer, providing a cathode layer, providing an electrolyte layer, comprise the step of providing an anode layer, a cathode layer, and an electrolyte layer with substantially similar mechanical properties.

8. The method of claim 6, wherein the step of compressing the cell stack, comprises the step of applying a rolling process to the cell stack to thin the layers of the cell stack.

9. The method of claim 7, wherein the step of providing an electrolyte layer, comprises the step of providing an electrolyte layer comprising an ionically conducting polymer having inert solids disposed therein.

10. A composite battery, comprising:

a plurality of cells, each cell comprising:

an anode layer having an extruding portion;

a cathode layer having an extruding portion ;

an electrolyte layer disposed to separate the anode and cathode layers;

the cells being successively stacked such that the extruding portions of the anode layers are in contact, and such that the extruding portions of the cathode layers are in contact.

FIG.1

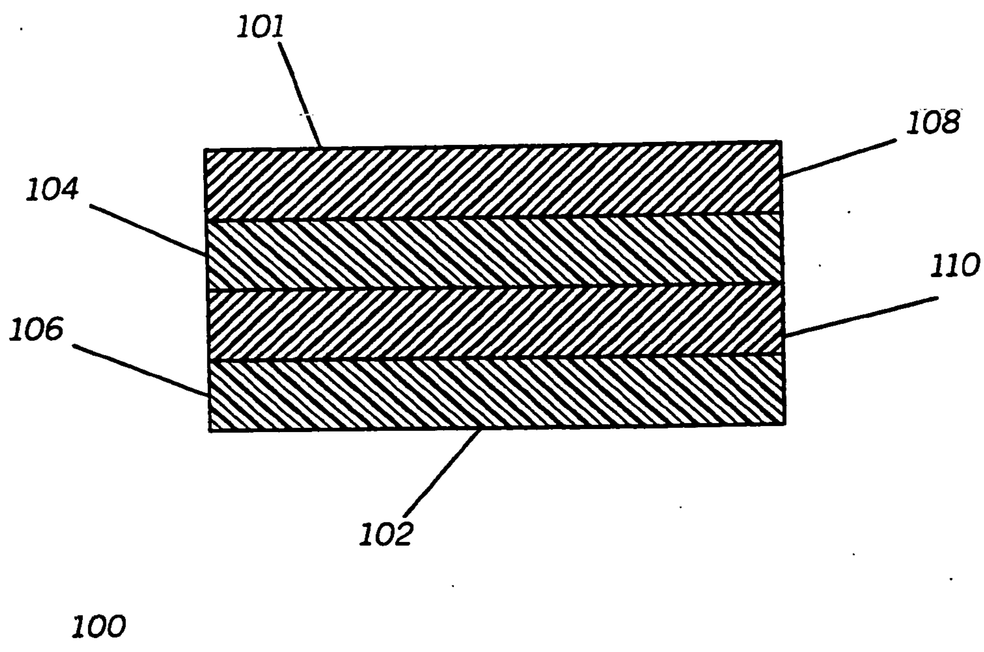


FIG.2

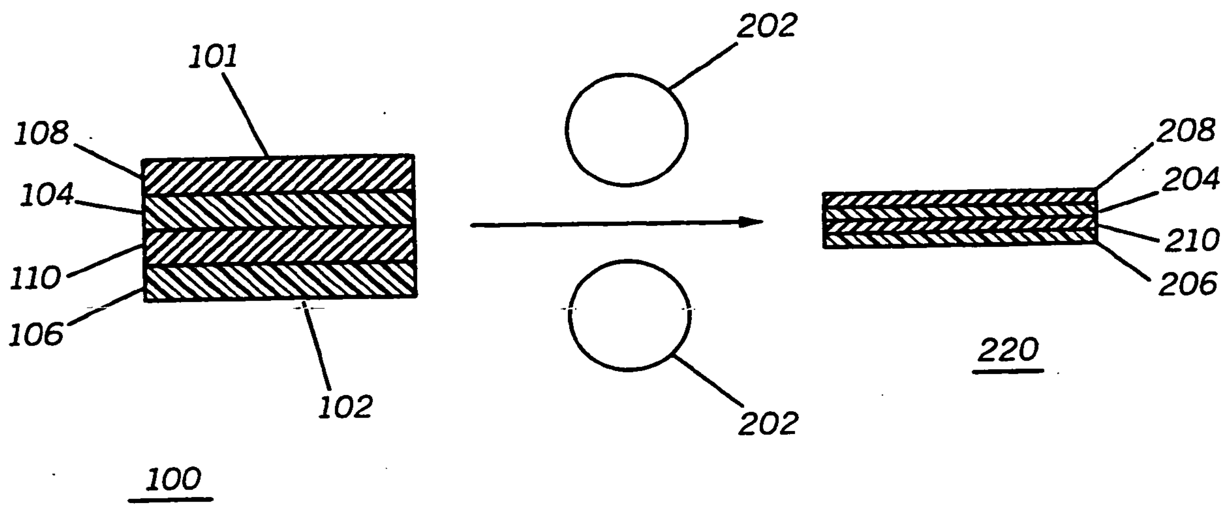

101
108
104
110
106
102
100
202
202
208
204
210
206
220

FIG.3

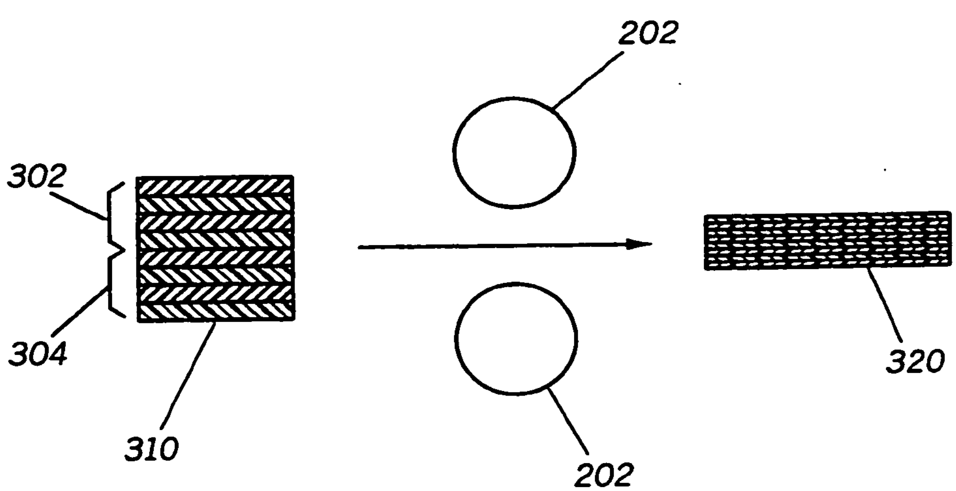

302
304
310
202
202
320

FIG.4

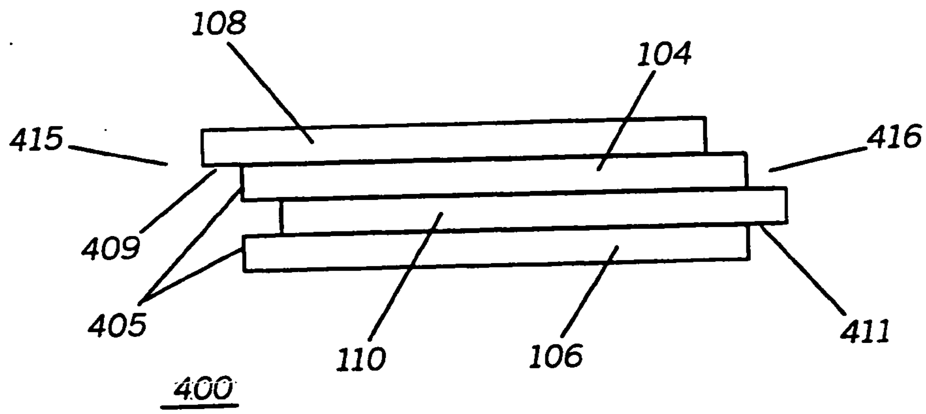


FIG.5

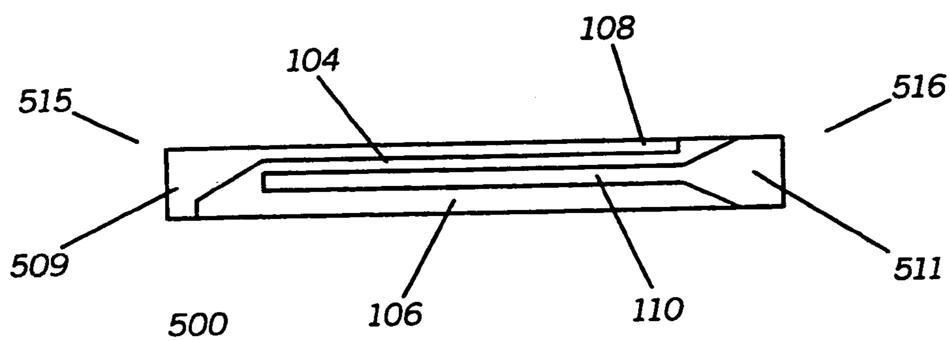
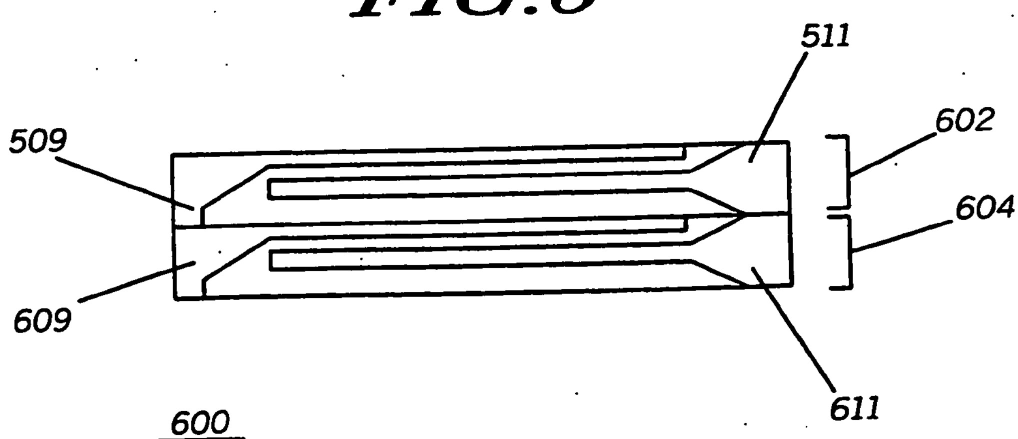


FIG.6



INTERNATIONAL SEARCH REPORT

International application No.
PCT/US95/07560

B. FIELDS SEARCHED
Electronic data bases consulted (Name of data base and where practicable terms used):

STN
search terms: composite battery, laminate, rollers, solid electrolyte